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(54) **Polypropylene moulded articles with increased -crystal content and bisamides as -nucleating agents.**

Polypropylenformkörper mit erhöhtem -Krystallitgehalt und Bisamidverbindungen als - Nukleierungsmittel

Articles moulés de polypropylène ayant une teneur en crystal élevée et bisamides en tant qu'agents de nucleation

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Description

[0001] The present invention relates to moulded articles obtained from a crystalline polypropylene resin composition containing an amide compound capable of acting as a nucleating agent for predominant formation of the β -crystal form and to a novel amide compound which is of use as a nucleating agent for the formation of the β -crystal form.

[0002] It is known that crystalline polypropylene may occur in α , β , γ and δ crystal forms as well as in the smectic crystal form which is formed on quenching of melted polypropylene. The β -crystal form (hereinafter referred to as " β -form") differs from the α -form which is found in the conventional natural pellet in that it is lower in melting point and in density, not to speak of differences in the mode of crystallization and of fracture, thus being of interest from application points of view (Kobunshi Kagaku 30, 694-698, (1973)).

[0003] For the production of crystalline polypropylene containing the β -form, a process comprising crystallizing molten polypropylene on a temperature gradient and a process comprising blending with a small amount of a nucleating agent (hereinafter referred to as " β -nucleating agent") are known. Since the former process is time-consuming and provides only a low yield, the latter process employing a β -nucleating agent is more advantageous for all practical purposes.

[0004] As such a β -nucleating agent, γ -quinacridone is well-known (Polymer Letters, 6, 539-546, (1968)). However, this nucleating agent has the drawback that it imparts a red tint to the product and requires special apparatus and operation for blending it with the polymer.

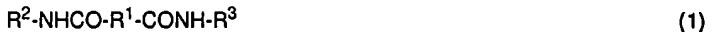
[0005] DE-A- 3206138 describes the use of the dianilide of adipic acid as nucleating agent for polypropylene, but does not discuss the structure of the crystals formed.

[0006] The object of the present invention is to provide a novel β -nucleating agent ensuring an efficient production of a product containing β -crystalline polypropylene in a large amount and much improved in the aspect of product color because of substantial absence of coloring property, and to provide a practically useful crystalline polypropylene resin composition containing said β -nucleating agent.

[0007] This object has been achieved by a class of amide compounds having a specific chemical structure. Some of said amide compounds are novel compounds which are not described before in the published literature.

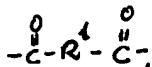
[0008] The moulded crystalline polypropylene resin composition of the present invention is characterized in that, in addition to a crystalline polypropylene resin, it contains at least one β -nucleating agent selected from amide compounds of the formula (1), amide compounds of the formula (2) and amide compounds of the formula (3) to be described below in an amount effective for increasing the content of the β -crystal form:

(1) an amide compound of the formula



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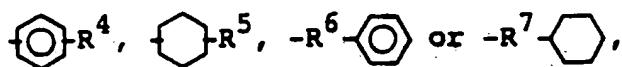
wherein



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is a residue of a C_{3-20} saturated or unsaturated aliphatic dicarboxylic acid, a residue of a C_{6-30} saturated or unsaturated alicyclic dicarboxylic acid or a residue of a C_{8-30} aromatic dicarboxylic acid; R^2 and R^3 are the same or different and each represents a C_{3-12} cycloalkyl group, a C_{3-12} cycloalkenyl group, or a group of the formula

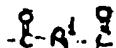
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wherein R^4 is a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} straight- or branched-chain alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; R^5 is a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} straight- or branched-chain alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; and R^6 and R^7 each represents a C_{1-4} straight- or branched-chain alkylene group; with the proviso that when R^4 is a C_{1-12} alkyl group or a C_{6-10} cycloalkyl group,

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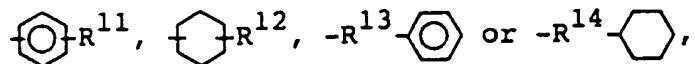


is a residue of a C₆ or C₈ saturated aliphatic dicarboxylic acid,

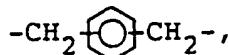
(2) an amide compound of the formula



5 wherein -NH-R⁸-NH- is a residue of a C₄-₂₈ alicyclic diamine, a residue of a C₄-₁₄ heterocyclic diamine or a residue of a C₆-₂₈ aromatic diamine; R⁹ and R¹⁰ are the same or different and each represents a C₃-₁₂ cycloalkyl group, a C₃-₁₂ cycloalkenyl group, or a group of the formula



15 wherein R¹¹ is a hydrogen atom, a C₁-₁₂ straight- or branched-chain alkyl group, a C₂-₁₂ straight- or branched-chain alkenyl group, a C₆-₁₀ cycloalkyl group or a phenyl group; R¹² is a C₁-₁₂ straight- or branched-chain alkyl group, a C₂-₁₂ alkenyl group, a C₆-₁₀ cycloalkyl group or a phenyl group; and R¹³ and R¹⁴ each represents a C₁-₄ straight- or branched-chain alkylene group; with the proviso that R⁸ is not



and

(3) an amide compound of the formula



30 wherein -NH-R¹⁵-CO- is a residue of a C₂-₂₉ saturated or unsaturated aliphatic amino acid, a residue of a C₇-₁₃ saturated or unsaturated alicyclic amino acid or a residue of a C₇-₁₅ aromatic amino acid; R¹⁶ and R¹⁷ are the same or different and R¹⁶ has the same meaning as R⁹ or R¹⁰ in the formula (2) and R¹⁷ has the same meaning as R² or R³ in the formula (1), which composition has a content of β -form crystals of 20-97% calculated by means of the following equation :

$$\beta\text{-form content (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta}) \quad (I)$$

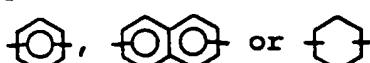
35 where A_α means the peak area of α -form and A_β means the peak area of β -form as determined by differential scanning calorimetry.

[0009] The present invention also provides the use of at least one amide compound of the above formulas (1), (2) and (3) as β -nucleating agents for polypropylene resin to achieve a proportion of the β -crystal form of 20 to 97% as calculated by means of equation (I).

[0010] Among these amide compounds of the formulas (1), (2) and (3), the compounds of the formula (1) and those of the formula (2) are preferred. Particularly preferred are amide compounds of the formula (1) wherein R¹ is -(CH₂)₄-.



50 and R² and R³ are the same or different and each represents a C₆-₈ cycloalkyl group or represents a phenyl group substituted by a C₁-₄ alkyl or cyclohexyl group, and amide compounds of the formula (2) wherein R⁸ is



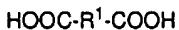
55 and R⁹ and R¹⁰ are the same or different and each is a cyclohexyl group or a phenyl group.

[0011] Processes for the production of amide compounds of the formulas (1), (2) and (3) are described below.

Amide compounds of the formula (1)

[0012] The amide compound of the formula (1) can be easily prepared by subjecting an aliphatic, alicyclic or aromatic dicarboxylic acid of the the formula

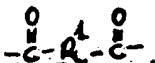
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(1a)

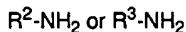
wherein

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is as defined hereinbefore to amidation reaction with an alicyclic or aromatic monoamine of the formula

15



wherein R^2 and R^3 are as defined hereinbefore.

[0013] This amidation reaction can be carried out in a conventional manner and any one of the following processes 20 may be mentioned as typical examples. Incidentally, where R^2 and R^3 in the formula (1) are dissimilar to each other, a substantially equimolar mixture of the corresponding two kinds of monoamines is employed.

(i) In an inert solvent, said dicarboxylic acid is reacted with said monoamine at a temperature of 60 to 200°C for 2 to 8 hours. The monoamine is generally used in an amount of 2 to 10 equivalents per one equivalent of the dicarboxylic acid. In this process, an activator is preferably used to accelerate the reaction. The activator that can be used includes phosphorus pentaoxide, polyphosphoric acid, phosphorus pentaoxide-methanesulfonic acid, phosphorous ester (e.g. triphenyl phosphite)-pyridine, phosphorous ester-metal salt (e.g. lithium chloride) or triphenylphosphine-hexachloroethane. Generally, about one mole of the activator is used per mole of the dicarboxylic acid.

(ii) In an inert solvent, dichloride of said dicarboxylic acid is reacted with said monoamine at a temperature of 0 to 100°C for 1 to 5 hours. The monoamine is used generally in an amount of 2 to 3 equivalents per one equivalent of the dicarboxylic acid dichloride.

(iii) In an inert solvent, a diester, particularly a di(C_{1-3} alkyl ester, of said dicarboxylic acid is reacted with said monoamine in the presence of a catalyst at 0-150°C for 3 to 10 hours. The monoamine is used generally in an amount of 2 to 20 equivalents per one equivalent of the dicarboxylic acid diester. The catalyst may be an acid or basic catalyst that is conventionally used in ester-amide interchange reactions, and is preferably a basic catalyst. Thus, there may be mentioned lithium, sodium, potassium; alkali metal hydrides such as lithium hydride, sodium hydride or potassium hydride; alkali metal hydroxides such as lithium hydroxide, sodium hydroxide or potassium hydroxide; metal alkoxides such as sodium methoxide, sodium ethoxide or potassium tert-butoxide; and alkali metal amides such as sodium amide or lithium diisopropylamide; among others. The catalyst is used generally in an equimolar amount relative to said dicarboxylic acid.

[0014] The inert solvent which can be used for the above processes (i), (ii) and (iii) includes, among others, benzene, toluene, xylene, chloroform, chlorobenzene, dichlorobenzene, tetrahydrofuran, dioxane, acetonitrile, N,N-dimethylformamide, N,N-dimethylacetamide and N-methylpyrrolidone.

[0015] As the dicarboxylic acid of the formula (1a) for use in said processes (i), (ii) and (iii), there may be mentioned the aliphatic, alicyclic or aromatic dicarboxylic acid corresponding to R^1 . Thus, R^1 is preferably a residue formed by elimination of the two carboxyl groups of one of the following aliphatic, alicyclic and aromatic dicarboxylic acids.

[0016] The aliphatic dicarboxylic acid specifically includes C_{3-20} , preferably C_{3-14} saturated or unsaturated aliphatic dicarboxylic acids, such as malonic acid, diphenylmalonic acid, succinic acid, phenylsuccinic acid, diphenylsuccinic acid, glutaric acid, 3,3-dimethylglutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebatic acid, 1,12-dodecanedioic acid, 1,14-tetradecanedioic acid or 1,18-octadecanedioic acid.

[0017] The alicyclic dicarboxylic acid specifically includes C_{6-30} , preferably C_{8-12} saturated or unsaturated alicyclic dicarboxylic acids such as 1,2-cyclohexane dicarboxylic acid, 1,4-cyclohexanedicarboxylic acid or 1,4-cyclohexane-diacetic acid.

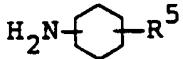
[0018] The aromatic dicarboxylic acid specifically includes C_{8-30} , preferably C_{8-22} aromatic dicarboxylic acids such as p-phenylenediacetic acid, p-phenylenediethanoic acid, phthalic acid, 4-tert-butylphthalic acid, isophthalic acid, 5-tert-butylisophthalic acid, terephthalic acid, 1,8-naphthalic acid, 1,4-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 3,3'-biphenyldicarboxylic acid, 4,4'-biphenyldicarbox-

5 ylic acid, 4,4'-binaphthylidicarboxylic acid, bis(3-carboxyphenyl)methane, bis(4-carboxyphenyl)methane, 2,2-bis(3-carboxyphenyl)propane, 2,2-bis(4-carboxyphenyl)propane, 3,3'-sulfonyldibenzoic acid, 4,4'-sulfonyldibenzoic acid, 3,3'-oxydibenzoic acid, 4,4'-oxydibenzoic acid, 3,3'-carbonyldibenzoic acid, 4,4'-carbonyldibenzoic acid, 3,3'-thiodibenzoic acid, 4,4'-thiodibenzoic acid, 4,4'-(p-phenylenedioxy)dibenzoic acid, 4,4'-isophthaloyldibenzoic acid, 4,4'-terephthaloyldibenzoic acid or dithiosalicylic acid.

[0019] On the other hand, the monoamine to be used in processes (i), (ii) and (iii) is the alicyclic or aromatic monoamine corresponding to R² or R³, i.e., R²-NH₂ or R³-NH₂.

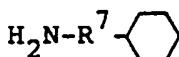
[0020] The alicyclic monoamine particularly includes C₃-C₁₂ cycloalkylamines, C₃-C₁₂ cycloalkenylamines,

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(wherein R⁵ is as defined above).

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20 (wherein R⁷ is as defined above) and the like, and specifically includes, among others, cyclopropylamine, cyclobutylamine, cyclopentylamine, cyclohexylamine, 2-methylcyclohexylamine, 3-methylcyclohexylamine, 4-methylcyclohexylamine, 2-ethylcyclohexylamine, 4-ethylcyclohexylamine, 2-propylcyclohexylamine, 2-isopropylcyclohexylamine, 4-propylcyclohexylamine, 4-isopropylcyclohexylamine, 2-tert-butylcyclohexylamine, 4-n-butylcyclohexylamine, 4-isobutylcyclohexylamine, 4-sec-butylcyclohexylamine, 4-tert-butylcyclohexylamine, 4-n-amylcyclohexylamine, 4-isoamylcyclohexylamine, 4-sec-amylcyclohexylamine, 4-tert-amylcyclohexylamine, 4-hexylcyclohexylamine, 4-heptylcyclohexylamine, 4-octylcyclohexylamine, 4-nonylcyclohexylamine, 4-decylcyclohexylamine, 4-undecylcyclohexylamine, 4-dodecylcyclohexylamine, 4-cyclohexylcyclohexylamine, 4-phenylcyclohexylamine, cycloheptylamine, cyclododecylamine, cyclohexylmethylamine, α -cyclohexylethylamine, β -cyclohexylethylamine, α -cyclohexylpropylamine, β -cyclohexylpropylamine or γ -cyclohexylpropylamine.

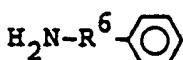
25 [0021] The aromatic monoamine particularly includes

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(wherein R⁴ is as defined above).

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45 (wherein R⁶ is as defined above) and the like, and specifically includes, among others, o-toluidine, m-toluidine, p-toluidine, o-ethylaniline, p-ethylaniline, o-propylaniline, m-propylaniline, p-propylaniline, o-cumidine, m-cumidine, p-cumidine, o-tert-butylaniline, p-n-butylaniline, p-isobutylaniline, p-sec-butylaniline, p-tert-butylaniline, p-n-amylaniline, p-isoamylaniline, p-sec-amylaniline, p-tert-amylaniline, p-hexylaniline, p-heptylaniline, p-octylaniline, p-nonylaniline, p-decylaniline, p-undecylaniline, p-dodecylaniline, p-cyclohexylaniline, o-aminodiphenyl, m-aminodiphenyl, p-aminodiphenyl, p-aminostyrene, benzylamine, α -phenylethylamine, β -phenylethylamine, α -phenylpropylamine, β -phenylpropylamine and γ -phenylpropylamine.

50 [0022] Among the amide compounds of the formula (1), the compounds which can be specifically represented by the following formula (4) are novel compounds not heretofore described in the literature.



55 wherein R¹⁸ means



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R^{19} and R^{20} may be the same or different and each means a C_{5-12} cycloalkyl group.

[0023] These novel amide compounds can be produced basically by the conventional amidation reactions and particularly by the above-described processes (i), (ii) and (iii). Thus, any one of the following methods can be used.

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(a) A naphthalenedicarboxylic acid or biphenyldicarboxylic acid of the formula $HOOC-R^{18}-COOH$ (hereinafter referred to as "dicarboxylic acid A") is reacted with 2 to 10 equivalents of a monoamine in an inert solvent at 60 to 200°C for 2 to 8 hours.

In order to accelerate this reaction, said activator is preferably employed.

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(b) A dichloride of said dicarboxylic acid A is reacted with 2 to 3 equivalents of a monoamine in an inert solvent at 0-100°C for 1 to 5 hours.

(c) A di(C_{1-3}) alkyl ester of dicarboxylic acid A is reacted with 2 to 20 equivalents of a monoamine in an inert solvent in the presence of an acid or basic catalyst, which is conventionally used in ester-amide interchange reactions as mentioned hereinbefore, and preferably in the presence of said basic catalyst, at 0 to 150°C for 3 to 10 hours.

20

[0024] The inert solvent which is employed in the above processes (a), (b) and (c) may be identical with the inert solvent mentioned hereinbefore for processes (i), (ii) and (iii).

[0025] The monoamine for use in the above processes is the monoamine corresponding to the formula $R^{19}-NH_2$ or $R^{20}-NH_2$. Particularly preferred are C_{5-12} cycloalkylamines such as cyclopentylamine, cyclohexylamine, cycloheptylamine, cyclooctylamine or cyclododecylamine.

[0026] The compounds obtained by the above processes (i), (ii) and (iii) or processes (a), (b) and (c) can each be isolated and purified by the conventional procedures such as chromatography, reprecipitation, recrystallization, fractional crystallization, and so on.

[0027] Among the amide compounds of the formula (1), those which are more effective include N,N' -dicyclohexylterephthalamide, N,N' -dicyclohexyl-2,6-naphthalenedicarboxamide, N,N' -dicyclooctyl-2,6-naphthalenedicarboxamide, N,N' -dicyclohexyl-1,4-cyclohexanedicarboxamide, N,N' -dicyclohexyl-4,4'-biphenyldicarboxamide, N,N' -bis(*p*-methylphenyl)hexanediamide, N,N' -bis(*p*-ethylphenyl)hexanediamide, N,N' -bis(4-cyclohexylphenyl)hexanediamide, N,N' -diphenylhexanediamide or N,N' -diphenyloctanediamide.

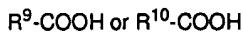
[0028] Among these, N,N' -dicyclohexylterephthalamide, N,N' -dicyclohexyl-2,6-naphthalenedicarboxamide and N,N' -dicyclohexyl-4,4'-biphenyldicarboxamide can provide/higher proportion of β -crystal form under quenching condition.

Amide compounds of the formula (2)

[0029] The amide compound of the formula (2) can be easily produced by subjecting an alicyclic, heterocyclic or aromatic diamine of the formula



45 wherein $-NH-R^8-NH-$ is as defined hereinbefore and an alicyclic or aromatic monocarboxylic acid of the formula



to amidation reaction in the conventional manner.

[0030] This amidation reaction can be conducted in various conventional ways, but typically any one of the following processes may be mentioned.

(i) The above-mentioned diamine is reacted with the monocarboxylic acid in an inert solvent at 60 to 200°C for 2 to 8 hours. The monocarboxylic acid is used generally in an amount of 2 to 10 equivalents per one equivalent of the diamine. In this process, too, the activator mentioned for process (i) is preferably employed in order to accelerate the reaction. The activator is used generally in an equimolar amount with respect to the diamine.

(ii) The above diamine is reacted with the acid chloride of said monocarboxylic acid in an inert solvent at 0 to 100°C for 1 to 5 hours. The monocarboxylic acid chloride is used generally in an amount of 2 to 3 equivalents per one

equivalent of the diamine.

(iii) The above diamine is reacted with an ester, particularly a C₁₋₃ alkyl ester, of said monocarboxylic acid in an inert solvent in the presence of a catalyst at 0 to 150°C for 3 to 10 hours. The monocarboxylic acid ester is generally used in an amount of 2 to 20 equivalents per one equivalent of the diamine. The catalyst may be selected from among the acid and basic catalysts for conventional ester-amide interchange reactions as mentioned for said process (iii) and is preferably a basic catalyst.

[0031] When R⁹ and R¹⁰ in the formula (2) are dissimilar, a substantially equimolar mixture of 2 kinds of corresponding monocarboxylic acids (or a substantially equimolar mixture of 2 kinds of corresponding monocarboxylic acid chlorides or a substantially equimolar mixture of 2 kinds of corresponding monocarboxylic acid esters) is employed.

[0032] The inert solvent for use in said processes (i'), (ii') and (iii') can be the same as the solvent mentioned for processes (i), (ii) and (iii) for production of the compound of the formula (1).

[0033] The compound obtained by the above processes can be isolated and purified by the conventional procedures such as chromatography, reprecipitation, recrystallization, fractional crystallization and so on.

[0034] In the above processes (i'), (ii') and (iii'), the diamine of the formula (2a) is the alicyclic, heterocyclic or aromatic diamine corresponding to R⁸. Thus, R⁸ is preferably a residue formed by elimination of the two amino groups of one of the following alicyclic, heterocyclic and aromatic diamines.

[0035] Thus, the alicyclic diamine includes C₄₋₂₈, preferably C₆₋₁₅ alicyclic diamines such as 1,2-diaminocyclohexane, 1,4-diaminocyclohexane, 4,4'-diaminodicyclohexyl, 4,4'-diamino-3,3'-dimethyldicyclohexyl, 4,4'-diaminodicyclohexylmethane, 4,4'-diamino-3,3'-dimethyldicyclohexylmethane, 1,3-bis(aminomethyl)cyclohexane, 1,4-bis(aminomethyl)cyclohexane, and additionally includes isophoronediamine or menthenediamine.

[0036] The heterocyclic diamine includes 5- or 6-membered heterocyclic diamines containing 1 or 2 nitrogen or sulfur atoms in the ring structure and having 4 to 14 carbon atoms, such as 2,3-diaminopyridine, 2,6-diaminopyridine, 3,4-diaminopyridine or o-tolidinesulfone.

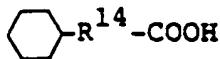
[0037] The aromatic diamine includes those containing 6 to 28, preferably 6 to 15 carbon atoms, such as o-phenylenediamine, m-phenylenediamine, p-phenylenediamine, 2,3-diaminotoluene, 2,4-diaminotoluene, 2,6-diaminotoluene, 3,4-diaminotoluene, 4,6-dimethyl-m-phenylenediamine, 2,5-dimethyl-p-phenylenediamine, 4,5-dimethyl-o-phenylenediamine, 2,4-diaminomesitylene, 1,5-diaminonaphthalene, 1,8-diaminonaphthalene, 2,3-diaminonaphthalene, 2,7-diaminonaphthalene, 9,10-diaminophenanthrene, 3,3',5,5'-tetramethylbenzidine, 3,3'-dimethyl-4,4'-diaminobiphenyl, 3,3'-dimethoxy-4,4'-diaminobiphenyl, 4,4'-diaminodiphenylmethane, 3,3'-diaminodiphenylmethane, 3,4'-diaminodiphenylmethane, 4,4'-methylenedi-o-toluidine, 4,4'-methylenedi-2,6-xylidine, 4,4'-methylenedi-2,6-diethylaniline, 4,4'-diamino-1,2-diphenylethane, 4,4'-diamino-2,2'-dimethylbibenzyl, 4,4'-diaminostilbene, 3,4'-diamino-2,2-diphenylpropane, 4,4'-diamino-2,2-diphenylpropane, 4,4'-diaminodiphenylether, 3,4'-diaminodiphenylether, 4,4'-thiodianiline, 2,2'-dithiodianiline, 4,4'-dithiodianiline, 3,3'-diaminodiphenylsulfone, 4,4'-diaminodiphenylsulfone, 3,3'-diaminobenzophenone, 4,4'-diaminobenzophenone, 4,4'-diaminobenzanilide, 2,7-diaminofluorene, 3,7-diamino-2-methoxyfluorene, bis-p-aminophenylaniline, 1,3-bis(4-aminophenylpropyl)benzene, 1,4-bis(4-aminophenylpropyl)benzene, 1,3-bis(4-aminophenoxy)benzene, 1,4-bis(4-aminophenoxy)benzene, 4,4'-bis(4-aminophenoxy)biphenyl, bis[4-(4-aminophenoxy)phenyl]ether, bis[4-(4-aminophenoxy)phenyl]sulfone or 9,9-bis(4-aminophenyl)fluorene.

[0038] As preferred examples of the alicyclic monocarboxylic acid represented by R⁹-COOH or R¹⁰-COOH, there may be mentioned C₄-C₁₃ cycloalkanecarboxylic acids, C₄-C₁₃ cycloalkenecarboxylic acids,



45

(wherein R¹² is as defined above),



50

(wherein R¹⁴ is as defined above), etc., and examples thereof are cyclopropanecarboxylic acid, cyclobutanecarboxylic acid, cyclopentanecarboxylic acid, 1-methylcyclopentanecarboxylic acid, 2-methylcyclopentanecarboxylic acid, 3-methylcyclopentanecarboxylic acid, 1-phenylcyclopentanecarboxylic acid, cyclopentenecarboxylic acid, cyclohexane-carboxylic acid, 1-methylcyclohexanecarboxylic acid, 2-methylcyclohexanecarboxylic acid, 3-methylcyclohexanecarboxylic acid, 4-methylcyclohexanecarboxylic acid, 4-propylcyclohexanecarboxylic acid, 4-butylcyclohexanecarboxylic acid, 4-pentylcyclohexanecarboxylic acid, 4-hexylcyclohexanecarboxylic acid, 4-phenylcyclohexanecarboxylic acid, 1-phenylcyclohexanecarboxylic acid, cyclohexenecarboxylic acid, 4-butylcyclohexenecarboxylic acid, cycloheptanecar-

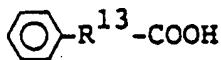
boxylic acid, 1-cycloheptenecarboxylic acid, 1-methylcycloheptanecarboxylic acid, 4-methylcycloheptanecarboxylic acid or cyclohexylacetic acid.

[0039] As preferred examples of the aromatic monocarboxylic acid represented by $R^9\text{-COOH}$ or $R^{10}\text{-COOH}$, there may be mentioned

5



10 (wherein R^{11} is as defined above),



15

(wherein R^{13} is as defined above), etc., and examples thereof are benzoic acid, o-methylbenzoic acid, m-methylbenzoic acid, p-methylbenzoic acid, p-ethylbenzoic acid, p-propylbenzoic acid, p-butylbenzoic acid, p-tert-butylbenzoic acid, p-pentylbenzoic acid, p-hexylbenzoic acid, o-phenylbenzoic acid, p-phenylbenzoic acid, p-cyclohexylbenzoic acid, phenylacetic acid, phenylpropionic acid or phenylbutyric acid.

20

[0040] The more desirable species of the diamide compound of the formula (2) which can be obtained from said diamine and monocarboxylic acid include N,N'-1,4-phenylenebiscyclohexanecarboxamide, N,N'-1,5-naphthalenebisbenzamide, N,N'-1,4-cyclohexanebisbenzamide or N,N'-1,4-cyclohexanebiscyclohexanecarboxamide.

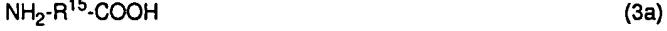
25

[0041] It should be noted that the desired effect cannot be obtained when the amide compound of the formula (2) is a compound synthesized by using xylylenediamine as the aromatic diamine. Such compounds of the formula (2) wherein R^8 is $-\text{CH}_2\text{-C}_6\text{H}_4\text{-CH}_2-$ are excluded from the scope of the present invention.

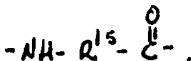
Amide compounds of the formula (3)

[0042] The amide compound of the formula (3) can be easily prepared by subjecting an aliphatic, alicyclic or aromatic amino acid of the formula (3a)

35



wherein



40 is as defined hereinbefore, a monocarboxylic acid chloride of the formula $R^{16}\text{-COCl}$ (wherein R^{16} is as defined hereinbefore) and a monoamine of the formula $R^{17}\text{-NH}_2$ (wherein R^{17} is as defined hereinbefore) to an amidation reaction.

[0043] This amidation reaction can be conducted, for example by reacting said amino acid of the formula (3a) with 1 to 2 equivalents of said monocarboxylic acid chloride in an inert solvent at 0 to 100°C for 1 to 5 hours, then adding 1 to 5 equivalents, based on the reaction product, of said monoamine and conducting the reaction, preferably in the presence of the activator mentioned for process (i), at a temperature of 60 to 200°C for 2 to 8 hours. The inert solvent may be any of the solvents mentioned hereinbefore in connection with process (i) for the production of said compound of the formula (1).

[0044] The compound obtained by the above process is purified by the conventional isolation and purification procedures such as chromatography, reprecipitation, recrystallization, fractional crystallization and so on.

50 [0045] The amino acid (3a) to be used in the above process is the aliphatic, alicyclic or aromatic amino acid corresponding to R^{15} . Thus, R^{15} is preferably a residue formed by elimination of one amino group and one carboxyl group from one of the aliphatic, alicyclic and aromatic amino acids mentioned below.

[0046] As preferred examples of said aliphatic amino acid, there may be mentioned C_{2-29} , more preferably C_{2-13} , saturated or unsaturated aliphatic amino acids such as aminoacetic acid, α -aminopropionic acid, β -aminopropionic acid, α -aminoacrylic acid, α -aminobutyric acid, β -aminobutyric acid, γ -aminobutyric acid, α -amino- α -methylbutyric acid, γ -amino- α -methylenebutyric acid, α -aminoisobutyric acid, β -aminoisobutyric acid, α -amino- α -valeric acid, δ -amino- α -valeric acid, β -aminocrotonic acid, α -amino- β -methylvaleric acid, α -aminoisovaleric acid, 2-amino-4-pentenoic acid, α -amino- α -caproic acid, 6-aminocaproic acid, α -aminoisocaproic acid, 7-aminoheptanoic acid, α -amino- α -caprylic acid,

8-aminocaprylic acid, 9-aminononanoic acid, 11-aminoundecanoic acid or 12-aminododecanoic acid.

[0047] The alicyclic amino acid includes C_{7-13} saturated or unsaturated alicyclic amino acids such as 1-aminocyclohexanecarboxylic acid, 2-aminocyclohexanecarboxylic acid, 3-aminocyclohexanecarboxylic acid, 4-aminocyclohexanecarboxylic acid, p-aminomethylcyclohexanecarboxylic acid or 2-amino-2-norbornanecarboxylic acid.

5 [0048] The aromatic amino acid specifically includes C_{7-15} aromatic amino acids such as α -aminophenylacetic acid, α -amino- β -phenylpropionic acid, 2-amino-2-phenylpropionic acid, 3-amino-3-phenylpropionic acid, α -aminocinnamic acid, 2-amino-4-phenylbutyric acid, 4-amino-3-phenylbutyric acid, anthranilic acid, m-aminobenzoic acid, p-aminobenzoic acid, 2-amino-4-methylbenzoic acid, 2-amino-6-methylbenzoic acid, 3-amino-4-methylbenzoic acid, 2-amino-3-methylbenzoic acid, 2-amino-5-methylbenzoic acid, 4-amino-2-methylbenzoic acid, 4-amino-3-methylbenzoic acid, 2-amino-3-methoxybenzoic acid, 3-amino-4-methoxybenzoic acid, 4-amino-2-methoxybenzoic acid, 4-amino-3-methoxybenzoic acid, 2-amino-4,5-dimethoxybenzoic acid, o-aminophenylacetic acid, m-aminophenylacetic acid, p-aminophenylacetic acid, 4-(4-aminophenyl)butyric acid, 4-aminomethylbenzoic acid, 4-aminomethylphenylacetic acid, o-aminocinnamic acid, m-aminocinnamic acid, p-aminocinnamic acid, p-aminohippuric acid, 2-amino-1-naphthoic acid, 3-amino-1-naphthoic acid, 4-amino-1-naphthoic acid, 5-amino-1-naphthoic acid, 6-amino-1-naphthoic acid, 7-amino-1-naphthoic acid, 8-amino-1-naphthoic acid, 1-amino-2-naphthoic acid, 3-amino-2-naphthoic acid, 4-amino-2-naphthoic acid, 5-amino-2-naphthoic acid, 6-amino-2-naphthoic acid, 7-amino-2-naphthoic acid or 8-amino-2-naphthoic acid.

10 [0049] The monoamine ($R^{17}-NH_2$) as a starting compound for the amide compound of the formula (3) is identical with the monoamine (R^2-NH_2 or R^3-NH_2) used as a starting compound for the amide compound of the formula (1), and similarly the monocarboxylic acid chloride ($R^{16}COCl$) is one derived from the monocarboxylic acid which is identical with the monocarboxylic acid (R^9COOH or $R^{10}COOH$) used as a starting compound for the amide compound of the formula (2).

15 [0050] Among various species of the amide compound of the formula (3), N-cyclohexyl-4-(N-cyclohexylcarboxylamino)benzamide and N-phenyl-5-(N-benzoylamino)pentanamide, for instance, are particularly effective.

20 [0051] The β -nucleating agent of the formula (1), (2) or (3) can be added to polypropylene resin at an optional stage, i.e. either during the polymerization reaction or after the polymer has been prepared.

25 [0052] The amount of said β -nucleating agent to be added is not critical insofar as the desired effect can be obtained. Generally, it is used in an amount effective to achieve a content of the β -crystal form of 20 to 97%. Especially, based on 100 parts by weight of polypropylene resin, 0.0001 to 5 parts by weight, preferably 0.001 to 1 part by weight, of the β -nucleating agent is employed. If the proportion of the β -nucleating agent is less than 0.0001 part by weight, formation of the β -crystal form may not be sufficient, while the use of the β -nucleating agent in excess of 5 parts by weight may not be rewarded with a commensurate effect and be uneconomical.

30 [0053] Thus, the β -nucleating agent of the invention is capable of causing a crystalline polypropylene resin to undergo transition to the β -crystal form at a very low level of addition and a molded product having a β -form content of 20 to 97%, particularly 40 to 97 %, more preferably 50 to 90 % can be obtained under the conventional molding conditions.

35 [0054] The term 'polypropylene resin' as used in this specification and claim means not only a polypropylene homopolymer but also a polymer composed predominantly of propylene, particularly a polymer composed of not less than 50% by weight, preferably not less than 80% by weight, of propylene. As examples of the latter polymer, there may be mentioned propylene-ethylene random copolymer, propylene-ethylene block copolymer, polymer blends of said 40 polypropylene resin with a small proportion of a thermoplastic resin, such as high-density polyethylene, polybutene-1 or poly-4-methylpentene-1.

45 [0055] The catalyst which can be used for the production of such polymers includes not only Ziegler-Natta catalyst which is commonly employed but also a combination catalyst, such as one wherein a transition metal compound (e.g. titanium halides such as titanium trichloride, titanium tetrachloride, etc.) supported on a support composed mainly of magnesium halide, such as magnesium chloride, is combined with an alkylaluminum compound (e.g. triethylaluminum, diethylaluminum chloride, etc.).

50 [0056] The melt flow rate (hereinafter referred to briefly as "MFR"; measured in accordance with JIS K 6758-1981) of the crystalline polypropylene resin can be appropriately selected according to the molding method to be employed and is generally 0.1 to 100 g/10 min. and preferably 0.5 to 50 g/10 min.

55 [0057] If required, the resin composition of the present invention may contain a variety of additives such as a stabilizer (e.g. epoxy compounds), an antioxidant (e.g. phenol compounds, phosphite compounds), an ultraviolet absorber (benzophenone compounds, benzotriazole compounds), a neutralizer, a nucleating agent, an antistatic agent, an anti-blocking agent, a lubricant (e.g. aliphatic hydrocarbons, higher fatty acids, and the alkali metal salts or alkaline earth metal salts thereof, fatty acid esters, higher fatty acid amides, rosin derivatives), a colorant, an elastomer, and a mineral (e.g. talc, hydrotalcite), each within a range not interfering with the effect of the invention.

[0058] The crystalline polypropylene resin composition is preferably produced by mixing said β -nucleating agent and polypropylene resin, with or without addition of said additives, in an ordinary mixer, such as a Henschel mixer, and if necessary, pelletizing the resulting mixture using an ordinary pelletizer, such as a single-screw extruder, in the per se

known manner.

[0059] The resulting crystalline polypropylene resin composition can be molded by various known techniques. Thus, injection molding, extrusion molding, compression molding and other molding techniques utilizing the conventional molding machines can be employed. Molding conditions may be those commonly employed. Typical preferred molding conditions may be as follows. Injection molding: resin temperature 200 to 300°C, preferably 240 to 280°C; mold temperature 30 to 120°C, preferably 50 to 80°C. Extrusion molding: resin temperature 200 to 300°C, preferably 240 to 280°C; chill roll temperature 40 to 140°C, preferably 60 to 120°C. Compression molding: temperature of melted resin about 200 to 300°C, preferably 240 to 280°C; cooling temperature 30 to 120°C, preferably 50 to 100°C.

[0060] Molded products of the invention, which contain much higher proportion of β -crystal form than before and which are satisfactory in the aspect of color, can be easily obtained by molding under the above-mentioned molding condition the resin composition prepared with use of the above-mentioned mixing method. Compared with the conventional polypropylene pellet which does not substantially contain β -crystals but is predominantly composed of α -crystals, the polypropylene molded product of the invention has lower melting point and requires less force for deformation under heating. Therefore, the molded products contribute a great deal to improved secondary processability and mechanical characteristics. The products encompass a wide variety of forms such as packages, sheets or films.

[0061] The ratio of α - to β -forms in the final product can be controlled as desired by suitably selecting molding conditions such as cooling conditions. For example, the proportion of β -form is increased as a higher cooling temperature is employed. Thus, it is possible to control the ratio of α - to β -forms by appropriately selecting cooling condition under the above molding conditions. This characteristic is beneficial particularly in the surface roughening of biaxially oriented film. The film having such a roughened surface displays excellent antiblocking property, printability and adhesion and is of great use in the fields of packaging film, printing paper, tracing paper and oil-immersion type plastic capacitors.

[0062] Moreover, the resin composition containing a naphthalenedicarboxylic acid di(C₃₋₁₂)cycloalkylamide, which is among the amide compounds of the invention, is extremely useful for improving the impact strength of moldings and can exhibit excellent utility in a variety of applications such as automotive and electrical parts.

25 Examples

[0063] The following examples and comparative examples are intended to describe the invention in further detail. In these examples and comparative examples, the β -form content, modulus in flexure and impact strength were determined by the following methods.

(1) Determination of β -form content

[0064] A sample (5-10 mg), cut from the sheet obtained in each Example and Comparative Example by punching, 35 is set in the sample holder of a differential scanning calorimeter (DSC) and melted by heating in a nitrogen gas atmosphere at 230°C for 5 minutes. The temperature is then decreased at a rate of 20°C/min to near room temperature and, then, increased again at a rate of 20°C/min. From the peak areas of the α - and β -crystal forms on the DSC thermogram thus obtained, the β -form content (area %) is calculated by means of the following equation.

$$40 \quad \beta\text{-form content (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta})$$

where A_{α} means the peak area of α -form and A_{β} means the peak area of β -form.

(2) Determination of modulus in flexure

[0065] The modulus was determined in accordance with JIS K 7203. The testing temperature was 25°C and the testing speed was 10 mm/min.

(3) Impact strength (duPont method)

[0066] In accordance with the falling weight impact test method described in JIS K 7211, the 50% destruction energy for a 2mm-thick sheet at 23°C was determined.

Example 1

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[0067] To 100 parts by weight of a propylene homopolymer powder (MFR = 14 g/10 min) was added 0.05 part by weight of N,N'-dicyclohexylterephthalamide and the mixture was milled in a Henschel mixer and pelletized with a 20 mm-diameter single-screw extruder. The resulting pellets were subjected to compression-molding by melting the pellets

at 230°C for 10 minutes and then placing it in a mold of 60°C and maintaining it therein for 5 minutes for solidification to give a 0.5 mm-thick sheet. The β -form content of the resulting sheet was 93% and the sheet showed no coloration.

Example 2

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[0068] Except that N,N'-dicyclohexyl-1,4-cyclohexanedicarboxamide was used as the β -nucleating agent, the procedure of Example 1 was otherwise repeated to provide a sheet. The β -form content of this sheet was 42% and the sheet showed no coloration.

10

Example 3

[0069] Except that N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 97% and the sheet showed no coloration.

15

Example 4

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[0070] Except that N,N'-dicyclohexyl-4,4'-biphenyldicarboxamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 59% and the sheet showed no coloration.

Example 5

25

[0071] Except that N,N'-bis(p-methylphenyl)hexanediamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 89% and the sheet showed no coloration.

Example 6

30

[0072] Except that N,N'-bis(p-ethylphenyl)hexanediamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 64% and the sheet showed no coloration.

Example 7

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[0073] Except that N,N'-bis(4-cyclohexylphenyl)hexanediamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 82% and the sheet showed no coloration.

40

Example 8

[0074] Except that 0.05 part by weight of N,N'-1,4-phenylenebis-cyclohexanecarboxamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 84% and the sheet showed no coloration.

45

Example 9

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[0075] Except that N,N'-1,5-naphthalenebisbenzamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 44% and the sheet showed no coloration.

Example 10

55

[0076] Except that N,N'-1,4-cyclohexanebisbenzamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 92% and the sheet showed no coloration.

Example 11

[0077] Except that N,N'-1,4-cyclohexanebiscyclohexanecarboxamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 85% and the sheet showed no coloration.

5

Example 12

[0078] Except that N-cyclohexyl-4-(N-cyclohexylcarbonylamino)benzamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 42% and the sheet showed no coloration.

10

Example 13

[0079] Except that N-phenyl-5-(N-benzoylamino)pentanamide was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 38% and the sheet showed no coloration.

15

Example 14

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[0080] Except that polypropylene homopolymer (MFR = 4.3 g/10 min) was used as the polypropylene, the procedure of Example 1 was repeated to provide a sheet. The β -form content of this sheet was 95% and the sheet showed no coloration.

25

Comparative Example 1

[0081] Except that N,N'-dicyclohexylterephthalamide was not added, the procedure of Example 1 was repeated to provide a sheet and its β -form content was determined. This sheet contained only a trace amount of β -form. The sheet showed no coloration.

30

Comparative Example 2

[0082] Except that γ -quinacridone was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. However, under the particular conditions used, the sheet contained only a trace amount of β -form. The sheet had a red color.

35

Example 15

[0083] To 100 parts by weight of a propylene homopolymer powder (MFR = 14 g/10 min) was added 0.2 part by weight of N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide and the mixture was milled in a Henschel mixer and pelletized with a 20 mm-diameter single-screw extruder. The resulting pellets were injection-molded at a resin temperature of 240°C and a mold temperature of 50°C to provide a test piece. The modulus in flexure of the above test piece was 1481 N/mm² (151 kgf/mm²). The duPont impact strength was 402 N · cm (41 kgf · cm).

40

Comparative Example 3

[0084] Except that N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide was not added, the procedure of Example 15 was otherwise repeated to provide a sheet. This sheet had a modulus in flexure of 1364 N/mm² (139 kgf/mm²) and a duPont impact strength of 29 N · cm (3 kgf · cm).

45

Example 16

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[0085] A 300 ml four-necked flask equipped with a stirrer, thermometer, condenser and gas inlet was charged with 6.48 g (0.03 mole) of 2,6-naphthalenedicarboxylic acid, 5.61 g (0.066 mole) of cyclopentylamine, 20.46 g (0.066 mol) of triphenyl phosphite, 25 g of pyridine and 100 g of N-methylpyrrolidone, and the reaction was carried out in a nitrogen gas atmosphere at 100°C for 3 hours, with stirring. After cooling, the reaction mixture was poured in 700 ml of isopropyl alcohol/water (1:1) for reprecipitation and washing. The mixture was stirred for 2 hours and the precipitate was then recovered by filtration and dried under reduced pressure at 110°C to provide 9.03 g (yield 86%) of N,N'-dicyclopentyl-

2,6-naphthalenedicarboxamide. This compound was a white powder melting at 375.4°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Example 17

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[0086] Except that 6.53 g (0.066 mole) of cyclohexylamine was used as the monoamine, the procedure of Example 16 was otherwise repeated to provide 9.64 g (yield 85%) of N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide. This compound was a white powder melting at 384.2°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

10

[0087] Except that the above compound was used as the β -nucleating agent, the procedure of Example 1 was otherwise repeated to provide a sheet. The β -form content of this sheet was 97% and the sheet showed no coloration.

Example 18

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[0088] Except that 8.38 g (0.066 mole) of cyclooctylamine was used as the monoamine, the procedure of Example 16 was otherwise repeated to provide 9.63 g (yield 74%) of N,N'-dicyclooctyl-2,6-naphthalenedicarboxamide. This compound was a white powder melting at 320.8°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

20

Example 19

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[0089] Except that 12.08 g (0.066 mole) of cyclododecylamine was used as the monoamine, the procedure of Example 16 was repeated to provide 13.3 g (yield 81%) of N,N'-dicyclododecyl-2,6-naphthalenedicarboxamide. This compound was a white powder melting at 321.8°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Example 20

30

[0090] Except that 6.48 g (0.03 mole) of 2,7-naphthalenedicarboxylic acid was used as the dicarboxylic acid, the procedure of Example 17 was otherwise repeated to provide 8.39 g (yield 74%) of N,N'-dicyclohexyl-2,7-naphthalenedicarboxamide. This compound was a white powder melting at 337.1°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Example 21

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[0091] Except that 7.26 g (0.03 mole) of 4,4'-biphenyldicarboxylic acid was used as the dicarboxylic acid, the procedure of Example 16 was otherwise repeated to provide 8.12 g (yield 72%) of N,N'-dicyclopentyl-4,4'-biphenyldicarboxamide. This compound was a white powder melting at 355.0°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

40

Example 22

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[0092] Except that 6.53 g (0.066 mole) of cyclohexylamine was used as the monoamine, the procedure of Example 21 was otherwise repeated to provide 9.94 g (yield 82%) of N,N'-dicyclohexyl-4,4'-biphenyldicarboxamide. This compound was a white powder melting at 370.8°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

[0093] Except that the above compound was used as the β -nucleating agent, the procedure of Example 1 was otherwise repeated to provide a sheet. The β -form content of this sheet was 59% and the sheet showed no coloration.

50

Example 23

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[0094] Except that 8.38 g (0.066 mole) of cyclooctylamine was used as the monoamine, the procedure of Example 21 was otherwise repeated to provide 10.8 g (yield 78%) of N,N'-dicyclooctyl-4,4'-biphenyldicarboxamide. This compound was a white powder melting at 320.2°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Example 24

[0095] Except that 12.08 g (0.066 mole) of cyclododecylamine was used as the monoamine, the procedure of Example 21 was otherwise repeated to provide 17.16 g (yield 82%) of N,N'-dicyclododecyl-4,4'-biphenyldicarboxamide.

5 This compound was a white powder melting at 346.7°C (decomp.). The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Example 25

10 [0096] A reactor was charged with 8.37 g (0.03 mole) of 2,2'-biphenyldicarboxylic acid dichloride, 11.88 g (0.12 mole) of cyclohexylamine and 120 g of chlorobenzene and the reaction was carried out in a nitrogen gas atmosphere at 80°C for 2.5 hours. After cooling, the reaction mixture was poured in 1000 ml of hexane for reprecipitation and washing. The precipitate was washed with 600 ml of a 0.5% aqueous sodium hydroxide solution and further with purified water thoroughly. The precipitate was collected by filtration and dried under reduced pressure at 110°C to provide 6.08 g (yield 50%) of N,N'-dicyclohexyl-2,2'-biphenyldicarboxamide. This compound was a white powder melting at 229.2-229.8°C. The elemental analysis and characteristic infrared absorptions are shown in Table 1.

Table 1

20	Example	Elemental Analysis (%)		Infrared spectra (cm ⁻¹)
		Calcd.	Found	
25	16	C:75.40 H: 7.48 N: 7.99	C:75.28 H: 7.61 N: 7.84	1330, 1553, 1628, 3264(amido) 1450, 2869, 2960(cyclopentyl) 697, 774, 838, 1595, 3074(2,6-naphthalene)
	17	C:76.16 H: 7.99 N: 7.40	C:76.04 H: 8.12 N: 7.28	1320, 1537, 1631, 3306(amido) 1450, 2853, 2937(cyclohexyl) 697, 778, 820, 1605, 3063(2,6-naphthalene)
	18	C:77.38 H: 8.81 N: 6.45	C:77.29 H: 8.88 N: 6.37	1315, 1529, 1632, 3317(amido) 1448, 2860, 2924(cyclooctyl) 697, 775, 819, 1605, 3062(2,6-naphthalene)
30	19	C:79.07 H: 9.95 N: 5.12	C:78.89 H:10.16 N: 4.96	1327, 1536, 1634, 3317(amido) 1469, 2862, 2944(cyclododecyl) 698, 768, 825, 1598, 3064(2,6-naphthalene)
	20	C:76.16 H: 7.99 N: 7.40	C:76.02 H: 8.10 N: 7.28	1338, 1565, 1640, 3241(amido) 1454, 2852, 2929(cyclohexyl) 729, 822, 855, 1620, 3068(2,7-naphthalene)
	21	C:76.56 H: 7.50 N: 7.44	C:76.38 H: 7.70 N: 7.32	1324, 1532, 1630, 3307(amido) 1451, 2869, 2957(cyclopentyl) 760, 843, 1492(4,4'-biphenyl)
35	22	C:77.19 H: 7.97 N: 6.92	C:77.03 H: 8.13 N: 6.77	1328, 1529, 1632, 3319(amido) 1449, 2854, 2936(cyclohexyl) 759, 843, 1491 (4,4'-biphenyl)
	23	C:78.22 H: 8.75 N: 6.08	C:78.10 H: 8.91 N: 5.92	1327, 1532, 1626, 3314(amido) 2847, 2926(cyclooctyl) 758,840,1493(4,4'-biphenyl)

Table 1 (continued)

Example	Elemental Analysis (%)		Infrared spectra (cm ⁻¹)
	Calcd.	Found	
24	C:79.67 H: 9.85 N: 4.89	C:79.41 H:10.00 N: 4.69	1329, 1538, 1636, 3324(amido) 1444, 2863, 2947(cyclododecyl) 757, 841, 1492(4,4'-biphenyl)
25	C:77.19 H: 7.97 N: 6.92	C:77.08 H: 8.06 N: 6.84	1339, 1558, 1635, 3241(amido) 1450, 2854, 2931(cyclohexyl) 758, 1471(2,2'-biphenyl)

15 [0097] Table 2 below shows the structure each of the β -nucleating agents used in Examples 1-25.

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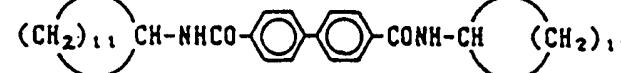
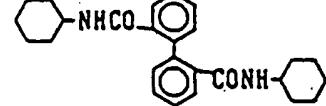
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Table 2

Example	Structure
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Table 2 (continued)

Example	Structure
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25	

Comparative Examples 4-9

[0098] Except that each of the amide compounds listed in Table 3 below (i.e., N,N'-diphenylbutanediamide; N,N'-diphenylpentanediamide, N,N'-diphenylheptanediamide, N,N'-diphenylnonanediamide, N,N'-diphenyldecanediamide and N,N'-diphenylterephthalamide) was used as the β -nucleating agent, the procedure of Example 1 was repeated to provide a sheet. The β -form contents of the resulting sheets are shown in Table 3 below.

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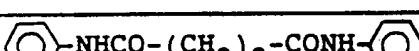
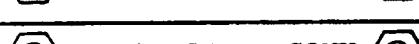
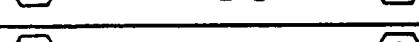
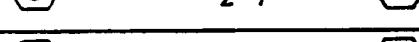
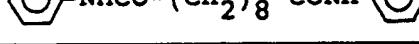
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Table 3

	Structure	β -form content (%)
Comparative Example 3/4		9
Comparative Example 4/5		3
Comparative Example 5/6		5
Comparative Example 6/7		trace
Comparative Example 7/8		trace
Comparative Example 8/9		trace

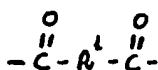
35 **Claims**

1. A molded crystalline polypropylene resin composition comprising a crystalline polypropylene resin and a β -nucleating agent, said β -nucleating agent being at least one selected from:

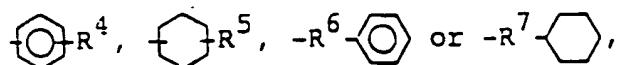
40 (1) an amide compound of the formula



wherein



50 represents a residue of a C₃₋₂₀ saturated or unsaturated aliphatic dicarboxylic acid, a residue of a C₆₋₃₀ saturated or unsaturated alicyclic dicarboxylic acid or a residue of a C₈₋₃₀ aromatic dicarboxylic acid; R² and R³ are the same or different and each represents a C₃₋₁₂ cycloalkyl group, a C₃₋₁₂ cycloalkenyl group, or a group of the formula

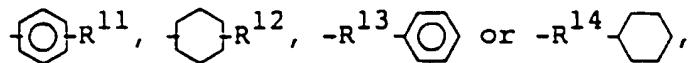


5 wherein R^4 represents a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} straight- or branched-chain alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; R^5 represents a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} straight- or branched-chain alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; and R^6 and R^7 each represents a C_{1-4} straight- or branched-chain alkylene group; with the proviso that when R^4 is a C_{1-12} alkyl group or a C_{6-10} cycloalkyl group, $-CO-R^1-CO-$ is a residue of a C_6 or C_8 saturated aliphatic dicarboxylic acid,

(2) an amide compound of the formula



10 wherein $-NH-R^8-NH-$ represents a residue of a C_{4-28} alicyclic diamine, a residue of a C_{4-14} heterocyclic diamine or a residue of a C_{6-28} aromatic diamine; R^9 and R^{10} are the same or different and each represents a C_{3-12} cycloalkyl group, a C_{3-12} cycloalkenyl group, or a group of the formula



20 wherein R^{11} represents a hydrogen atom, a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} straight- or branched-chain alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; R^{12} represents a C_{1-12} straight- or branched-chain alkyl group, a C_{2-12} alkenyl group, a C_{6-10} cycloalkyl group or a phenyl group; R^{13} and R^{14} each represents a C_{1-4} straight- or branched-chain alkylene group; with the proviso that R^8 is not



and

(3) an amide compound of the formula



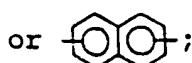
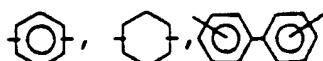
30 wherein $-NH-R^{15}-CO-$ represents a residue of a C_{2-29} saturated or unsaturated aliphatic amino acid, a residue of a C_{7-13} saturated or unsaturated alicyclic amino acid or a residue of a C_{7-15} aromatic amino acid; and R^{16} and R^{17} are the same or different and R^{16} has the same meaning as R^9 or R^{10} in the formula (2) and R^{17} has the same meaning as R^2 or R^3 in the formula (1) which composition has a content of β -form crystals of 20 to 97% calculated by means of the following equation

$$\beta\text{-form content (\%)} = 100 \times A_\beta / (A_\alpha + A_\beta) \quad (I)$$

40 where A_α means the peak area of α -form and A_β means the peak area of β -form determined by differential scanning calorimetry.

45 2. The molded composition as claimed in claim 1 wherein the β -nucleating agent is an amide compound of the formula (1).

3. The molded composition as claimed in claim 2 wherein R^1 is $-(CH_2)_4-$.



R² and R³ are the same or different and each represents a C₆-8 cycloalkyl group or represents a phenyl group substituted by a C₁₋₄ alkyl or cyclohexyl group.

4. The molded composition as claimed in claim 2 wherein the β -nucleating agent is at least one amide compound selected from N,N'-dicyclohexylterephthalamide, N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide, N,N'-dicyclooctyl-2,6-naphthalenedicarboxamide, N,N'-dicyclohexyl-1,4-cyclohexanedicarboxamide, N,N'-dicyclohexyl-4,4'-biphenyldicarboxamide, N,N'-bis(p-methylphenyl)-hexanediamide, N,N'-bis(p-ethylphenyl)hexanediamide and N,N'-bis(4-cyclohexylphenyl)hexanediamide.

10 5. The molded composition as claimed in claim 2 wherein the β -nucleating agent is at least one amide compound selected from N,N'-dicyclohexylterephthalamide, N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide and N,N'-dicyclohexyl-4,4'-biphenyldicarboxamide.

15 6. The molded composition as claimed in claim 1 wherein the β -nucleating agent is an amide compound of the formula (2).

7. The molded composition as claimed in claim 6 wherein R⁸ is



R⁹ and R¹⁰ are the same or different and each is a cyclohexyl group or a phenyl group.

25 8. The molded composition as claimed in claim 6 wherein the β -nucleating agent is at least one compound selected from N,N'-1,4-phenylenebis-cyclohexanecarboxamide, N,N'-1,5-naphthalenebis-benzamide, N,N'-1,4-cyclohexanebis-benzamide and N,N'-1,4-cyclohexanebis-cyclohexanecarboxamide.

30 9. The molded composition as claimed in claim 1 wherein the β -nucleating agent is an amide compound of the formula (3).

10. The molded composition as claimed in claim 9 wherein the β -nucleating agent is at least one compound selected from N-cyclohexyl-4-(N-cyclohexylcarbonylamino)benzamide and N-phenyl-5-(N-benzoylamino)pentanamide.

35 11. Use of at least one amide compound of the formulae (1), (2) and (3) of any of claims 1 to 10 as β -nucleating agent for polypropylene resin to achieve a content of β -form crystals of 20 to 97% calculated by means of equation (I)

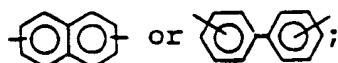
$$\beta\text{-form content (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta}) \quad (I)$$

40 where A _{α} means the peak area of α -form and A _{β} means the peak area of β -form determined by differential scanning calorimetry.

45 12. An amide compound of the formula



50 wherein R¹⁸ represents



55 and R¹⁹ and R²⁰ are the same or different and each represents a cycloalkyl group of 5 to 12 carbon atoms.

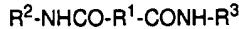
Patentansprüche

1. Geformte kristalline Polypropylenharzmasse, umfassend ein kristallines Polypropylenharz und einen β -Keimbild-

ner, wobei der β -Keimbildner mindestens einer ist, ausgewählt aus:

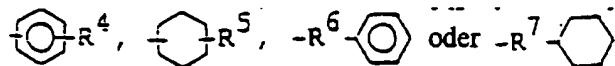
(1) einer Amidverbindung der Formel

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in der $\text{-CO-R}^1\text{-CO-}$ einen Rest einer gesättigten oder ungesättigten aliphatischen C_{3-20} -Dicarbonsäure, einen Rest einer gesättigten oder ungesättigten alicyclischen C_{6-30} -Dicarbonsäure oder einen Rest einer aromatischen C_{8-30} -Dicarbonsäure darstellt; R^2 und R^3 gleich oder verschieden sind und jeweils einen C_{3-12} -Cycloalkylrest, einen C_{3-12} -Cycloalkenylrest oder einen Rest der Formel



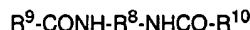
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darstellen, wobei R^4 einen linearen oder verzweigten C_{1-12} -Alkylrest, einen linearen oder verzweigten C_{2-12} -Alkenylrest, einen C_{6-10} -Cycloalkylrest oder eine Phenylgruppe darstellt; R^5 einen linearen oder verzweigten C_{1-12} -Alkylrest, einen linearen oder verzweigten C_{2-12} -Alkenylrest, einen C_{6-10} -Cycloalkylrest oder eine Phenylgruppe darstellt; und R^6 und R^7 jeweils einen linearen oder verzweigten C_{1-4} -Alkylenrest darstellen; mit der Maßgabe, daß, wenn R^4 ein C_{1-12} -Alkylrest oder ein C_{6-10} -Cycloalkylrest ist, $\text{-CO-R}^1\text{-CO-}$ ein Rest einer gesättigten aliphatischen C_6 - oder C_8 -Dicarbonsäure ist,

(2) einer Amidverbindung der Formel

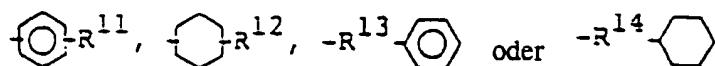
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in der $\text{-NH-R}^8\text{-NH-}$ einen Rest eines alicyclischen C_{4-28} -Diamins, einen Rest eines heterocyclischen C_{4-14} -Diamins oder einen Rest eines aromatischen C_{6-28} -Diamins darstellt; R^9 und R^{10} gleich oder verschieden sind und jeweils einen C_{3-12} -Cycloalkylrest, einen C_{3-12} -Cycloalkenylrest oder einen Rest der Formel

35



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darstellt,

wobei R^{11} ein Wasserstoffatom, einen linearen oder verzweigten C_{1-12} -Alkylrest, einen linearen oder verzweigten C_{2-12} -Alkenylrest, einen C_{6-10} -Cycloalkylrest oder eine Phenylgruppe darstellt; R^{12} einen linearen oder verzweigten C_{1-12} -Alkylrest, einen C_{2-12} -Alkenylrest, einen C_{6-10} -Cycloalkylrest oder eine Phenylgruppe darstellt; R^{13} und R^{14} jeweils einen linearen oder verzweigten C_{1-4} -Alkylenrest darstellen; mit der Maßgabe, daß R^8 nicht

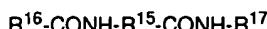
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ist, und

(3) einer Amidverbindung der Formel

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in der $\text{-NH-R}^{15}\text{-CO-}$ einen Rest einer gesättigten oder ungesättigten aliphatischen C_{2-29} -Aminosäure, einen Rest einer gesättigten oder ungesättigten alicyclischen C_{7-13} -Aminosäure oder einen Rest einer aromatischen C_{7-15} -Aminosäure darstellt; und R^{16} und R^{17} gleich oder verschieden sind und R^{16} die gleiche Bedeutung wie R^9 oder R^{10} in der Formel (2) hat und R^{17} die gleiche Bedeutung wie R^2 oder R^3 in der Formel (1) hat, wobei die Masse einen Gehalt an Kristallen der β -Form von 20 bis 97 %, berechnet mittels folgender Gleichung aufweist:

$$\text{Gehalt an } \beta\text{-Form (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta}) \quad (I)$$

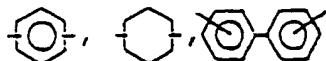
wobei A_α die Peakfläche der α -Form bedeutet und A_β die Peakfläche der β -Form bedeutet, die mit Differenzscanningkalorimetrie bestimmt werden.

2. Geformte Masse nach Anspruch 1, in der der β -Keimbildner eine Amidverbindung der Formel (1) ist.

5

3. Geformte Masse nach Anspruch 2, in der R^1 $-(CH_2)_4-$.

10



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darstellt; R^2 und R^3 gleich oder verschieden sind und jeweils einen C_{6-8} -Cycloalkylrest oder eine mit einem C_{1-4} -Alkylrest oder einer Cyclohexylgruppe substituierte Phenylgruppe darstellen.

25

4. Geformte Masse nach Anspruch 2, in der der β -Keimbildner mindestens eine Amidverbindung ist, ausgewählt aus N,N'-Dicyclohexylterephthalamid, N,N'-Dicyclohexyl-2,6-naphthalindicarbonsäureamid, N,N'-Dicyclooctyl-2,6-naphthalindicarbonsäureamid, N,N'-Dicyclohexyl-1,4-cyclohexandicarbonsäureamid, N,N'-Dicyclohexyl-4,4'-biphenyldicarbonsäureamid, N,N'-Bis(p-methylphenyl)hexandiamid, N,N'-Bis(p-ethylphenyl)hexandiamid und N,N'-Bis(4-cyclohexylphenyl)hexandiamid.

20

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5. Geformte Masse nach Anspruch 2, in der der β -Keimbildner mindestens eine Amidverbindung ist, ausgewählt aus N,N'-Dicyclohexylterephthalamid, N,N'-Dicyclohexyl-2,6-naphthalindicarbonsäureamid und N,N'-Dicyclohexyl-4,4'-biphenyldicarbonsäureamid.

35

6. Geformte Masse nach Anspruch 1, in der der β -Keimbildner eine Amidverbindung der Formel (2) ist.

35

7. Geformte Masse nach Anspruch 6, in der R^8

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ist; R^9 und R^{10} gleich oder verschieden sind und jeweils eine Cyclohexylgruppe oder eine Phenylgruppe sind.

50

8. Geformte Masse nach Anspruch 6, in der der β -Keimbildner mindestens eine Verbindung ist, ausgewählt aus N,N'-1,4-Phenylbiscyclohexancarbonsäureamid, N,N'-1,5-Naphthalinbisbenzamid, N,N'-1,4-Cyclohexanbisbenzamid und N,N'-1,4-Cyclohexanbiscyclohexancarbonsäureamid.

55

9. Geformte Masse nach Anspruch 1, in der der β -Keimbildner eine Amidverbindung der Formel (3) ist.

10. Geformte Masse nach Anspruch 9, in der der β -Keimbildner mindestens eine Verbindung ist, ausgewählt aus N-Cyclohexyl-4-(N-cyclohexylcarbonylamino)benzamid und N-Phenyl-5-(N-benzoylamino)pentanamid.

11. Verwendung mindestens einer Amidverbindung der Formeln (1), (2) und (3) nach einem der Ansprüche 1 bis 10 als β -Keimbildner für Polypropylenharz, um einen Gehalt an β -Formkristallen von 20 bis 97 % zu erreichen, berechnet mittels Gleichung (I)

$$5 \quad \text{Gehalt an } \beta\text{-Form (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta}) \quad (1)$$

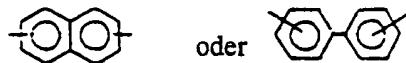
wobei A_α die Peakfläche der α -Form bedeutet und A_β die Peakfläche der β -Form bedeutet, die mit Differentialscaningkalorimetrie bestimmt werden.

10 12. Amidverbindung der Formel



in der R¹⁸

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20 darstellt; und R^{19} und R^{20} gleich oder verschieden sind und jeweils einen Cycloalkylrest mit 5 bis 12 Kohlenstoff-
atomen darstellen.

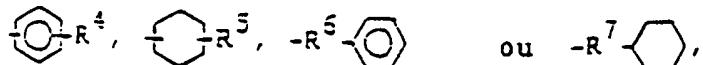
Revendications

25 1. Composition moulée de résine de polypropylène cristalline comprenant une résine de polypropylène cristalline et un agent de β -nucléation, ledit agent de β -nucléation étant au moins un agent choisi parmi:

(1) un composé amide de formule :



dans laquelle $-C(=O)-R^1-C(=O)-$ représente un résidu d'un acide dicarboxylique aliphatique saturé ou insaturé en C_{3-20} , un résidu d'un acide dicarboxylique alicyclique saturé ou insaturé en C_{6-30} ou un résidu d'acide dicarboxylique aromatique en C_{8-30} ; R^2 et R^3 sont identiques ou différents et représentent chacun un groupe cycloalkyle en C_{3-12} , un groupe cycloalcényle en C_{3-12} ou un groupe de formule



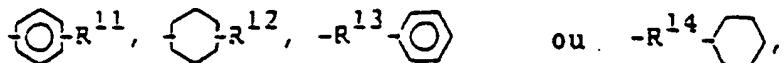
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où R^4 représente un groupe alkyle en C_{1-12} à chaîne droite ou ramifiée, un groupe alcényle en C_{2-12} à chaîne droite ou ramifiée, un groupe cycloalkyle en C_{6-10} ou un groupe phényle; R^5 représente un groupe alkyle en C_{1-12} à chaîne droite ou ramifiée, un groupe alcényle en C_{2-12} à chaîne droite ou ramifiée, un groupe cycloalkyle en C_{6-10} ou un groupe phényle; et R^6 et R^7 représentent chacun un groupe alkylène en C_{1-4} à chaîne droite ou ramifiée; à condition que lorsque R^4 est un groupe alkyle en C_{1-12} ou un groupe cycloalkyle en C_{6-10} , $-CO-R^1-CO-$ soit un résidu d'un acide dicarboxylique aliphatique saturé en C_6 ou C_8 ,
(2) un composé amide de formule :

15

$$B^9\text{-CONH-}B^8\text{-NHCO-}B^{10}$$

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où R¹¹ représente un atome d'hydrogène, un groupe alkyle en C₁₋₁₂ à chaîne droite ou ramifiée, un groupe alcényle en C₂₋₁₂ à chaîne droite ou ramifiée, un groupe cycloalkyle en C₆₋₁₀ ou un groupe phényle ; R¹² représente un groupe alkyle en C₁₋₁₂ à chaîne droite ou ramifiée, un groupe alcényle en C₂₋₁₂, un groupe cycloalkyle en C₆₋₁₀ ou un groupe phényle ; R¹³ et R¹⁴ représentent chacun un groupe alkylène en C₁₋₄ à chaîne droite ou ramifiée ; à condition que R⁸ ne soit pas

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(3) un composé amide de formule :

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dans laquelle -NH-R¹⁵-CO- représente un résidu d'un acide aminé aliphatique saturé ou insaturé en C₂₋₂₉, un résidu d'un acide aminé alicyclique saturé ou insaturé en C₇₋₁₃ ou un résidu d'un acide aminé aromatique en C₇₋₁₅ ; et R¹⁶ et R¹⁷ sont identiques ou différents et R¹⁶ a la même signification que R⁹ ou R¹⁰ dans la formule (2) et R¹⁷ a la même signification que R² ou R³ dans la formule (1).

laquelle composition a une teneur en cristaux de forme β de 20 à 97% calculée à l'aide de l'équation suivante:

$$\text{teneur en forme } \beta (\%) = 100 \times A_\beta / (A_\alpha + A_\beta) \quad (I)$$

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dans laquelle A_α représente la surface du pic de forme α et A_β représente la surface du pic de forme β déterminées par calorimétrie différentielle à balayage.

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2. Composition moulée suivant la revendication 1, dans laquelle l'agent de β-nucléation est un composé amide de formule (1).
3. Composition moulée suivant la revendication 2, dans laquelle R¹ est -(CH₂)₄-.

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R² et R³ sont identiques ou différents et représentent chacun un groupe cycloalkyle en C₆₋₈ ou représente un groupe phényle substitué par un groupe alkyle en C₁₋₄ ou un groupe cyclohexyle.

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4. Composition moulée suivant la revendication 2, dans laquelle l'agent de β-nucléation est au moins un composé amide choisi parmi le N,N'-dicyclohexyltéraphthalamide, le N,N'-dicyclohexyl-2,6-naphtalènedicarboxamide, le N,N'-dicyclooctyl-2,6-naphtalènedicarboxamide, le N,N'-dicyclohexyl-1,4-cyclohexanedicarboxamide, le N,N'-dicyclohexyl-4,4'-biphényldicarboxamide, le N,N'-bis-(p-méthylphényle)hexanediamide, le N,N'-bis-(p-éthylphényle)-hexanediamide et le N,N'-bis-(4-cyclohexylphényle)-hexanediamide.
5. Composition moulée suivant la revendication 2, dans laquelle l'agent de β-nucléation est au moins un composé amide choisi parmi le N,N'-dicyclohexyltéraphthalamide, le N,N'-dicyclohexyl-2,6-naphtalènedicarboxamide et le N,N'-dicyclohexyl-4,4'-biphényldicarboxamide.
6. Composition moulée suivant la revendication 1, dans laquelle l'agent de β-nucléation est un composé amide de formule (2).
7. Composition moulée suivant la revendication 6, dans laquelle R⁸ est



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R^9 et R^{10} sont identiques ou différents et sont chacun un groupe cyclohexyle ou un groupe phényle.

8. Composition moulée suivant la revendication 6, dans laquelle l'agent de β -nucléation est au moins un composé choisi parmi le N,N'-1,4-phénylenebis-cyclohexanecarboxamide, le N,N'-1,5-naphtalènebis-benzamide, le N,N'-1,4-cyclohexanebisbenzamide et le N,N'-1,4-cyclohexanebiscyclohexanecarboxamide.

10 9. Composition moulée suivant la revendication 1, dans laquelle l'agent de β -nucléation est un composé amide de formule (3).

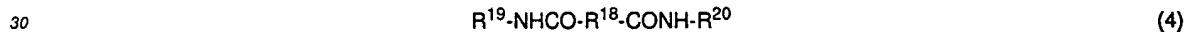
15 10. Composition moulée suivant la revendication 9, dans laquelle l'agent de β -nucléation est au moins un composé choisi parmi le N-cyclohexyl-4-(N-cyclohexylcarbonylamino)-benzamide et le N-phényl-5-(N-benzoylamino)-penta-

20 11. Utilisation d'au moins un composé amide des formules (1), (2) et (3) suivant l'une quelconque des revendications 1 à 10 en tant qu'agent de β -nucléation pour une résine de polypropylène pour obtenir une teneur en cristaux de forme β de 20 à 97% calculée à l'aide de l'équation (I):

$$\text{teneur en forme } \beta \text{ (\%)} = 100 \times A_{\beta} / (A_{\alpha} + A_{\beta}) \quad (I)$$

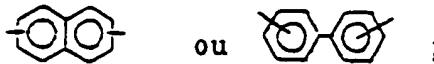
25 dans laquelle A_{α} représente la surface du pic de forme α et A_{β} représente la surface du pic de forme β déterminées par calorimétrie différentielle à balayage.

12. Composé amide de formule



dans laquelle R^{18} représente

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et R^{19} et R^{20} sont identiques ou différents et représentent chacun un groupe cycloalkyle de 5 à 12 atomes de carbone.

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